FORM PTO-1390	U S DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE	ATTORNEY'S DOCKET NUMBER
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371		4197-113
		U S APPLICATION NO (If known, see 37 CFR 1 5) 10/088773

INTERNATIONAL APPLICATION NO	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED
PCT/DE00/03411	29 September 2000	15 October 1999

TITLE OF INVENTION
METHOD AND DEVICE FOR CONTINUALLY PRODUCING AN EXTRUSION SOLUTION

APPLICANT(S) FOR DO/EO/US
BAUER, Ralf-Uwe and KIND, Uwe

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S C 371
3. ☐ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).
4. ☒ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.

5. ☒ A copy of the International Application as filed (35 U.S C. 371(c)(2))
   a. ☒ is transmitted herewith (required only if not transmitted by the International Bureau).
   b. ☒ has been transmitted by the International Bureau.
   c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).

6. ☒ A translation of the International Application into English (35 U S C 371(c)(2)).

7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S C 371(c)(3))
   a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
   b. ☐ have been transmitted by the International Bureau.
   c. ☐ have not been made, however, the time limit for making such amendments has NOT expired
   d. ☒ have not been made and will not be made

8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S C 371(c)(3))

9. ☒ An oath or declaration of the inventor(s) (35 U S C 371(c)(4)) ***(Unsigned)**

10. ☒ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5))

Items 11. to 16. below concern other document(s) or information included:

11. ☐ An Information Disclosure Statement under 37 CFR 1 97 and 1 98

12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3 28 and 3 31 is included.

13. ☒ A FIRST preliminary amendment
    ☐ A SECOND or SUBSEQUENT preliminary amendment

14. ☐ A substitute specification

15. ☒ A small entity statement.

16. ☐ Other items or information:

NOTE: This application is being filed with an unsigned Oath or Declaration under the provisions of 37 CFR § 1.53 in order that applicants may secure a filing date of March 18, 2002. Upon receipt of a "Notice to File Missing Parts - Filing Date Granted," an executed Declaration and Power of Attorney, will be filed in the Patent and Trademark Office. The undersigned agent affirmatively states that she has been duly authorized and appointed to file this application on behalf of the applicants and that the Declaration and Power of Attorney to be filed hereafter will confirm the undersigned agent's authorization and appointment. Applicants are entitled to small entity status within the meaning of 37 CFR § 1.9.

10/088773

JC18 Rec'd PCT/PTO 18 MAR 2002

17. ☒ The following fees are submitted:	CALCULATIONS	PTO USE ONLY
Basic National Fee (37 CFR 1 492(a)(1)-(5)): Search Report has been prepared by the EPO or JPO $860.00		
International preliminary examination fee paid to USPTO (37 CFR 1 482) $0.00		
No International preliminary examination fee paid to USPTO (37 CFR 1 482) but international search fee paid to USPTO (37 CFR 1 445(a)(2)) $0.00		
Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO $1000.00		
International preliminary examination fee paid to USPTO (37 CFR 1 482) and all claims satisfied provisions of PCT Article 33(2)-(4) $0 00		
ENTER APPROPRIATE BASIC FEE AMOUNT =	$ 860.00	
Surcharge of **$130.00** for furnishing the oath or declaration later than ☐ 20 ☐ 30 months from the earliest claimed priority date (37 CFR 1.492(e)).	$	

Claims	Number Filed	Number Extra	Rate		
Total Claims	15 -20 =	0	X **$18.00**	$	
Independent Claims	3- 3 =	0	X **$80.00**	$	
Multiple dependent claim(s) (if applicable)			+ **$270.00**	$	
TOTAL OF ABOVE CALCULATIONS =				860.00	
Reduction by 1/2 for filing by small entity, if applicable Verified Small Entity statement must also be filed. (Note 37 CFR 1 9, 1 27, 1 28)				$ 430.00	
SUBTOTAL =				$ 430.00	
Processing fee of **$130.00** for furnishing the English translation later than ☐ 20 ☐ 30 Months from the earliest claimed priority date (37 CFR 1 492(f)) +				$	
TOTAL NATIONAL FEE =				$ 430.00	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3 28, 3 31) **$40.00** per property +				$	
TOTAL FEE ENCLOSED =				$ 430.00	
				Amount to be: refunded	$
				Charged	$

a. ☒ A check in the amount of $430.00 to cover the above fees is enclosed.

b. ☐ Please charge my Deposit Account No _____ in the amount of $_____ to cover the above fees. A duplicate copy of this sheet is enclosed.

c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 08-3284. A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not yet been met, a petition to revive (37 CFR 1.127(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

Steven J. Hultquist
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MARIANNE FUIERER
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4197-113
PATENT APPLICATION

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:	**BAUER, et al.**
Application No.:	New U.S. National Stage Application of PCT International Application No. PCT/DE00/03411
International Filing Date:	29 September 2000
Priority Date Claimed:	15 October 1999 (German Appl. No. 199 49 720.0)
U.S. National Phase Filing Date:	Date of mailing identified below
Title:	**METHOD AND DEVICE FOR CONTINUALLY PRODUCING AN EXTRUSION SOLUTION**

EXPRESS MAIL CERTIFICATE

I hereby certify that I am mailing the attached documents to the Commissioner for Patents on the date specified, in an envelope addressed to the Commissioner for Patents, Box Patent Application, Washington, DC 20231, and Express Mailed under the provisions of 37 CFR 1 10

Katrina Holland
Name of Person Mailing This Document

Katrina Holland
Signature

March 18, 2002
Date

EV037733018US
Express Mail Label Number

PRELIMINARY AMENDMENT

Commissioner for Patents
BOX PATENT APPLICATION
Washington, D.C. 20231

Sir:

Prior to examination of the above-identified new national phase patent application, please amend the application, as follows:

In the Specification [1]

Please insert on page 1, between the title of the application and the first paragraph, the following new paragraph:

CROSS-REFERENCE TO RELATED APPLICATIONS

This application is filed under the provisions of 35 U. S.C. §371 and claims the priority of International Patent Application No. PCT/DE00/03411 filed September 29, 2000, which in turn claims priority of German Patent Application No. 199 49 720.6 filed October 15, 1999.

In the Claims

Please amend claims 1-11 to read as follows:

1. A method for the continuous production of an extrusion solution for the formation of cellulosic molded bodies, such as fibers and films, according to the lyocell method, comprising:

   (a) forming a cellulose suspension comprising pulp and an aqueous phase in a mass ratio in the range from 1:3 to 1:40 and shearing the cellulose suspension for a period of time in the range from 5 to 200 minutes;

[1] Applicants has provided a marked-up version of amended paragraph, and claims 1-11 in Appendix A, and a clean set of all pending claims, amended to date, in Appendix B

(b) dewatering the cellulose suspension to form a material with a cellulose content in the range from 20 to 80 mass-percent, wherein the aqueous phase from the dewatering is at least partially recycled for subsequent use in step (a);

(c) conveying the damp cellulose material through a first shear zone in the absence of N-methylmorpholine-N-oxide, wherein the damp cellulose material is homogenized in the first shear zone;

(d) adding a sufficient amount of aqueous N-methylmorpholine-N-oxide to the homogenized cellulose material to form a cellulose suspension with a content of N-methylmorpholine-N-oxide in the liquid phase in a range from 70 to 80 mass-percent and conveying the homogenized suspension through a second shear zone with the cellulose material essentially completely filling up the available conveyor cross-section in the shear zones; and

(e) converting the cellulose suspension in aqueous N-methylmorpholine-N-oxide into the extrusion solution by evaporating water evaporation with shearing.

2. The method according to Claim 1, wherein part of the aqueous phase from step (b) is combined with fresh water for formation of the cellulose suspension in step (a).

3. The method according to Claim 1, wherein the aqueous phase used in step (a) contains dissolved components.

4. The method according to Claim 1, wherein the cellulose suspension is dewatered in step (b) with the aid of vacuum and/or pressure into a fleece and the water content of the fleece is determined with the aid of an infrared moisture measurer and is used to regulate the predetermined pressure parameter and/or the addition of the aqueous N-methylmorpholine-N-oxide in step (d).

5. The method according to Claim 1, wherein the formation of the cellulose solution in step (e) is performed in a strong shear field with small heat exchange surfaces up to an NMMO/H_2O mol ratio in the range from 1:0.8 to 1:1.2.

6. The method according to Claim 1, wherein the cellulose is enzymatically activated in step (a) by treating the cellulose suspension with 0.01 to 10 mass-percent enzyme, in relation to cellulose, at a temperature in the range between 20 and 70 °C and a pH value in the range from 3 to 10 for a duration in the range from 0.1 to 10 hours.

7. The method according to Claim 6, wherein the enzymatic treatment is performed with 0.1 to 3.0 mass-percent enzyme at 30 to 60 °C and a pH value of 4.5 to 8 for a duration of 0.5 to 2 hours.

8. The method according to Claim 1, wherein steps (a) and (b) are performed in the pulp factory.

9. A device for the continuous production of an extrusion solution for the formation of cellulosic molded bodies, such as fibers and films, according to the lyocell method, comprising:

a mixing tank with suspending elements;
a pulp supply connecting piece for introducing pulp into the mixing tank;
an aqueous suspension agent supply connecting piece for introducing an aqueous suspension agent into the mixing tank;
a separating apparatus for partial dewatering a formed aqueous pulp suspension;
a drain connecting piece communicatively connected to and between the mixing tank and the separating apparatus;
a return line connected to the separating apparatus for returning any aqueous suspension agent separated from the separating apparatus to the aqueous supply connecting piece of the mixing tank;

a shearing apparatus including a first homogenization zone and adjoining suspending zone, having a first feed connecting piece for introducing pulp fleece from the separating apparatus at the beginning of the homogenization zone, a second feed connecting piece at the beginning of the suspending zone for introducing solvent, and a drain connecting piece for removing suspension at the end of the suspending zone; and

a concentration and dissolving apparatus communicatively connected at a first end to the drain connecting piece of the shearing apparatus, a solution outlet connecting piece at the other end of the dissolving apparatus, and at least one vapor outlet connecting piece.

10. The device according to Claim 9, wherein the separating apparatus is a vacuum screen belt press.

11. The device according to Claim 9, wherein the separating apparatus is a vacuum screen drum filter.

Please add the following claims.

12. The method according to Claim 1 wherein the cellulose is enzymatically activated between steps (a) and (b), by treating the cellulose suspension with 0.01 to 10 mass-percent enzyme, in relation to cellulose, at a temperature in the range between 20 and 70 °C and a pH value in the range from 3 to 10 for a duration in the range from 0.1 to 10 hours.

13. The method according to claim 6, wherein the enzyme is a cellulase.

14. The method according to claim 12, wherein the enzyme is a cellulase.

15. A method for the continuous production of an extrusion solution for the formation of cellulosic molded bodies, such as fibers and films, according to the lyocell method, wherein

(a) forming a cellulose suspension comprising pulp and an aqueous phase in a mass ratio in the range from 1:3 to 1:40 and shearing the cellulose suspension for a period of time in the range from 5 to 200 minutes;

(b) dewatering the cellulose suspension to form a fleece material with a cellulose content in the range from 20 to 80 mass-percent:

(c) shearing the fleece material in the absence of N-methylmorpholine-N-oxide, to form a homogenized cellulose material;

(d) adding a sufficient amount of aqueous N-methylmorpholine-N-oxide to the homogenized cellulose material to form a cellulose suspension with a content of N-methylmorpholine-N-oxide in the liquid phase in a range from 70 to 80 mass-percent and shearing the cellulose suspension in aqueous N-methylmorpholine-N-oxide to evaporate excess water and form the extrusion solution.

REMARKS

It is requested that the examination and prosecution of this application proceed on the basis of these amended and new claims 1-15.

Respectfully submitted,

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APPENDIX A

In the Specification

Please insert on page 1, between the title of the application and the first paragraph, the following new paragraph:

CROSS-REFERENCE TO RELATED APPLICATIONS

This application is filed under the provisions of 35 U. S.C. §371 and claims the priority of International Patent Application No. PCT/DE00/03411 filed September 29, 2000, which in turn claims priority of German Patent Application No. 199 49 720.6 filed October 15, 1999.

In the Claims

Please amend claims 1-11 to read as follows:

1. A method for the continuous production of an extrusion solution for the formation of cellulosic molded bodies, such as fibers and films, according to the lyocell method, comprising: [wherein]

   (a) forming a cellulose suspension [is formed]comprising [from] pulp and an aqueous phase in a mass ratio in the range from 1:3 to 1:40 and shearing the cellulose suspension [maintained] for a period of time in the range from 5 to 200 minutes;[with shearing,]

   (b) dewatering the cellulose suspension [is dewatered] to form a material with a cellulose content in the range from 20 to 80 mass-percent, wherein [and] the aqueous phase from the dewatering [resulting in this case] is at least partially recycled for subsequent use in step (a);[,]

(c) conveying the damp cellulose material [is conveyed, with homogenization,] through a first shear zone in the absence of N-methylmorpholine-N-oxide, wherein the damp cellulose material is homogenized in the first shear zone;

(d) adding a sufficient amount of aqueous N-methylmorpholine-N-oxide to the homogenized cellulose material to form a cellulose suspension with a content of N-methylmorpholine-N-oxide in the liquid phase in a range from 70 to 80 mass-percent and conveying the homogenized suspension [the homogenized cellulose material is conveyed] through a second shear zone [after the addition of sufficient aqueous N-methylmorpholine-N-oxide that after the mixing a suspension with a content of N-methylmorpholine-N-oxide in the liquid phase in a range from 70 to 80 mass-percent results,] with the cellulose material essentially completely filling up the available conveyor cross-section in the shear zones [essentially completely,]; and

(e) converting the cellulose suspension in aqueous N-methylmorpholine-N-oxide [formed is converted] into the extrusion solution by evaporating water evaporation with shearing.

2. The method according to Claim 1, [characterized in that] wherein part of [partly] the aqueous phase from step (b) is combined with [and partly] fresh water [is used] for formation of the cellulose suspension in step (a).

3. The method according to Claim 1 [or 2],wherein [characterized in that] the aqueous phase used in step (a) [an aqueous phase is used which may] contains dissolved components.

4. The method according to Claim 1, [one of Claims 1 to 3,

characterized in that] wherein the cellulose suspension is dewatered in step (b) with the aid of vacuum and/or pressure into a fleece and the water content of the fleece is determined with the aid of an infrared moisture measurer and is used to regulate the predetermined pressure parameter and/or the addition of the aqueous N-methylmorpholine-N-oxide in step (d).

5. The method according to Claim 1, wherein [any one of Claims 1 to 4, characterized in that in step (e)] the formation of the cellulose solution in step (e) is performed in a strong shear field with small heat exchange surfaces up to an NMMO/H_2O mol ratio in the range from 1:0.8 to 1:1.2.

6. The method according to Claim 1 [one of Claims 1 to 5, characterized in that] wherein the cellulose is enzymatically activated in step (a) [or between steps (a) and (b),] by treating the cellulose suspension with 0.01 to 10 mass-percent enzyme, in relation to cellulose, at a temperature in the range between 20 and 70 °C and a pH value in the range from 3 to 10 for a duration in the range from 0.1 to 10 hours.

7. The method according to Claim 6, [characterized in that] wherein the enzymatic treatment is performed with 0.1 to 3.0 mass-percent enzyme at 30 to 60 °C and a pH value of 4.5 to 8 for a duration of 0.5 to 2 hours.

8. The method according to Claim 1, wherein [one of Claims 1 to 7, characterized in that] steps (a) and (b) are performed in the pulp factory.

9. A device for the continuous production of an extrusion solution for the formation of cellulosic molded bodies, such as fibers and films, according to the lyocell method, comprising: [performing the method according one of Claims 1 to 8, having]

a mixing tank [(1)] with suspending [organs,]elements; a pulp supply connecting piece[s (3, 2)] for introducing pulp into the mixing tank; an aqueous suspension agent supply connecting piece for introducing an [and] aqueous suspension agent into the mixing tank;[, and] a separating apparatus for partial dewatering a formed aqueous pulp suspension; a drain connecting piece [(6)] communicatively connected to and between the mixing tank and the separating apparatus; [to the suspension from,

a separating apparatus (8) connected with the drain connecting piece (6) for partial separation of the suspension agent from the cellulose,]

a return line [(4)] connected to the separating apparatus for returning any aqueous suspension agent separated [suspension agent] from the separating apparatus [(8)] to [a] the aqueous supply connecting piece [(2)] of the mixing tank; [(1) with a discard line (9) for the possible partial suspension agent discard,]

a shearing apparatus [(11),] including a first homogenization zone and [an] adjoining suspending zone, having a first feed connecting piece [(12)] for introducing pulp fleece from the separating apparatus [(8)] at the beginning of the homogenization zone, a second feed connecting piece [(15)] at the beginning of the suspending zone for introducing solvent, and a drain connecting piece [(17)] for removing suspension at the end of the suspending zone; and

a concentration and dissolving apparatus [(18)] communicatively connected at a first end [having a feed connecting piece connected] to the drain connecting piece [(17)] of the shearing apparatus [(11) at one end], a solution outlet connecting piece [(19)] at the other end of the dissolving apparatus, and at least one vapor outlet connecting piece [(20)].

10. The device according to Claim 9, wherein [characterized in that] the separating apparatus [(8)] is a vacuum screen belt press.

11. The device according to Claim 9, wherein [characterized in that] the separating apparatus [(8)] is a vacuum screen drum filter.

APPENDIX B

In the Specification

Please insert on page 1, between the title of the application and the first paragraph, the following new paragraph:

CROSS-REFERENCE TO RELATED APPLICATIONS

This application is filed under the provisions of 35 U. S.C. §371 and claims the priority of International Patent Application No. PCT/DE00/03411 filed September 29, 2000, which in turn claims priority of German Patent Application No. 199 49 720.6 filed October 15, 1999.

Pending Claims 1-15

1. A method for the continuous production of an extrusion solution for the formation of cellulosic molded bodies, such as fibers and films, according to the lyocell method, comprising:

   (a) forming a cellulose suspension comprising pulp and an aqueous phase in a mass ratio in the range from 1:3 to 1:40 and shearing the cellulose suspension for a period of time in the range from 5 to 200 minutes;

   (b) dewatering the cellulose suspension to form a material with a cellulose content in the range from 20 to 80 mass-percent, wherein the aqueous phase from the dewatering is at least partially recycled for subsequent use in step (a);

   (c) conveying the damp cellulose material through a first shear zone in the absence of N-methylmorpholine-N-oxide, wherein the damp cellulose material is homogenized in the first shear zone;

(d) adding a sufficient amount of aqueous N-methylmorpholine-N-oxide to the homogenized cellulose material to form a cellulose suspension with a content of N-methylmorpholine-N-oxide in the liquid phase in a range from 70 to 80 mass-percent and conveying the homogenized suspension through a second shear zone with the cellulose material essentially completely filling up the available conveyor cross-section in the shear zones; and

(e) converting the cellulose suspension in aqueous N-methylmorpholine-N-oxide into the extrusion solution by evaporating water evaporation with shearing.

2. The method according to Claim 1, wherein part of the aqueous phase from step (b) is combined with fresh water for formation of the cellulose suspension in step (a).

3. The method according to Claim 1, wherein the aqueous phase used in step (a) contains dissolved components.

4. The method according to Claim 1, wherein the cellulose suspension is dewatered in step (b) with the aid of vacuum and/or pressure into a fleece and the water content of the fleece is determined with the aid of an infrared moisture measurer and is used to regulate the predetermined pressure parameter and/or the addition of the aqueous N-methylmorpholine-N-oxide in step (d).

5. The method according to Claim 1, wherein the formation of the cellulose solution in step (e) is performed in a strong shear field with small heat exchange surfaces up to an NMMO/H_2O mol ratio in the range from 1:0.8 to 1:1.2.

6. The method according to Claim 1, wherein the cellulose is enzymatically activated in step (a) by treating the cellulose suspension with 0.01 to 10 mass-percent enzyme, in

relation to cellulose, at a temperature in the range between 20 and 70 °C and a pH value in the range from 3 to 10 for a duration in the range from 0.1 to 10 hours.

7. The method according to Claim 6, wherein the enzymatic treatment is performed with 0.1 to 3.0 mass-percent enzyme at 30 to 60 °C and a pH value of 4.5 to 8 for a duration of 0.5 to 2 hours.

8. The method according to Claim 1, wherein steps (a) and (b) are performed in the pulp factory.

9. A device for the continuous production of an extrusion solution for the formation of cellulosic molded bodies, such as fibers and films, according to the lyocell method, comprising:

a mixing tank with suspending elements;
a pulp supply connecting piece for introducing pulp into the mixing tank;
an aqueous suspension agent supply connecting piece for introducing an aqueous suspension agent into the mixing tank;
a separating apparatus for partial dewatering a formed aqueous pulp suspension;
a drain connecting piece communicatively connected to and between the mixing tank and the separating apparatus;
a return line connected to the separating apparatus for returning any aqueous suspension agent separated from the separating apparatus to the aqueous supply connecting piece of the mixing tank;
a shearing apparatus including a first homogenization zone and adjoining suspending zone, having a first feed connecting piece for introducing pulp fleece from the separating apparatus at the beginning of the homogenization zone, a second feed connecting piece at the beginning of the suspending zone for introducing solvent, and a drain connecting piece for removing suspension at the end of the suspending zone; and

a concentration and dissolving apparatus communicatively connected at a first end to the drain connecting piece of the shearing apparatus, a solution outlet connecting piece at the other end of the dissolving apparatus, and at least one vapor outlet connecting piece.

10. The device according to Claim 9, wherein the separating apparatus is a vacuum screen belt press.

11. The device according to Claim 9, wherein the separating apparatus is a vacuum screen drum filter.

Please add the following claims.

12. The method according to Claim 1 wherein the cellulose is enzymatically activated between steps (a) and (b), by treating the cellulose suspension with 0.01 to 10 mass-percent enzyme, in relation to cellulose, at a temperature in the range between 20 and 70 °C and a pH value in the range from 3 to 10 for a duration in the range from 0.1 to 10 hours.

13. The method according to claim 6, wherein the enzyme is a cellulase.

14. The method according to claim 12, wherein the enzyme is a cellulase.

15. A method for the continuous production of an extrusion solution for the formation of cellulosic molded bodies, such as fibers and films, according to the lyocell method, wherein

(a) forming a cellulose suspension comprising pulp and an aqueous phase in a mass ratio in the range from 1:3 to 1:40 and shearing the cellulose suspension for a period of time in the range from 5 to 200 minutes;

(b) dewatering the cellulose suspension to form a fleece material with a cellulose content in the range from 20 to 80 mass-percent:

(c) shearing the fleece material in the absence of N-methylmorpholine-N-oxide, to form a homogenized cellulose material;

(d) adding a sufficient amount of aqueous N-methylmorpholine-N-oxide to the homogenized cellulose material to form a cellulose suspension with a content of N-methylmorpholine-N-oxide in the liquid phase in a range from 70 to 80 mass-percent and shearing the cellulose suspension in aqueous N-methylmorpholine-N-oxide to evaporate excess water and form the extrusion solution.

1/prt

METHOD AND DEVICE FOR CONTINUOUS PRODUCTION OF AN EXTRUSION SOLUTION

The present invention relates to a method for the continuous production of an extrusion solution for the formation of cellulose molded bodies, such as fibers and films, according to the lyocell method. The present invention also relates to a device for performing the method.

A discontinuous method for producing a suspension of cellulose in aqueous aminoxide is known from WO 94/28217. In this case, shredded cellulose and an aminoxide solution are mixed in a horizontal mixing chamber by a rotor having radial stirring elements. Twenty-one minutes is given as the duration for one batch. This mode of operation is disadvantageous because, due to the continuous feeding of the subsequent dissolving stage, two such mixing chambers have to be operated. In addition, the complete emptying of the mixing chambers is connected with difficulties.

A semicontinuous method for producing a cellulose solution is known from WO 96/33302. In this case, first a suspension of the cellulose in aminoxide solution is formed, from which water is evaporated before the production of the cellulose solution in a separate apparatus. It is disadvantageous in this case that the cellulose activation in the aqueous aminoxide solution is limited and a separate thermal step is necessary for concentration. The soluble components of the pulp reach the spinning solution and may lead to disadvantageous properties of the cellulose products.

Furthermore, producing a cellulose suspension in aqueous N-methylmorpholine-N-oxide (NMMO) by mixing the shredded cellulose directly in an annular layer mixer with the aqueous, e.g. 75 mass-percent NMMO is known from WO 96/33221. The suspension produced is brought to solution in a separate Filmtruder. It is disadvantageous in the annular layer mixer that only shredded, essentially dry cellulose may be used. If the cellulose contains water, the layer formation in the mixer and the mixing with the separately added NMMO solution is made more difficult. The water must be thermally separated in this case as well. The same disadvantages exist as in the method described in WO 96/33302. Since the suspension is transported as a layer, the throughput related to the apparatus cross-section is low.

Suspending the pulp before the formation of a homogeneous suspension in aminoxide solution in water and separating it again partially from the suspension agent after a certain time is known from German Patent 198 37 210.8. Recirculation of the suspension agent is not described.

The present invention has the object of providing a method and a device for the continuous production of an extrusion solution for the formation of cellulosic molded bodies according to the lyocell method, in which the pulp used is activated so that its solubility and speed of dissolving is elevated. In addition, soluble impurities of the pulp are to be partially separated in the method, so that their transition into the extrusion solution is reduced. Furthermore, a method for the continuous production of an extrusion solution for the lyocell method is to be provided which is distinguished by reduced consumption of thermal energy for water separation and by reduced thermal stress of the aminoxide and cellulose. Further advantages result from the following description.

These objects are achieved according to the present invention with the method initially described in that

(a) a cellulose suspension is formed from pulp and an aqueous phase in a mass ratio in the range from 1:3 to 1:40 and maintained for a period of time in the range from 5 to 200 minutes with shearing,

(b) the cellulose suspension is dewatered to form a material with a cellulose content in the range from 20 to 80 mass-percent and the aqueous phase resulting in this case is at least partially recycled in step (a),

(c) the damp cellulose material is conveyed, with homogenization, through a first shear zone in the absence of N-methylmorpholine-N-oxide,

(d) the homogenized cellulose material is conveyed through a second shear zone after the addition of enough aqueous N-methylmorpholine-N-oxide that after mixing a suspension with a content of N-methylmorpholine-N-oxide in the liquid phase in a range from 70 to 80 mass-percent results, and

(e) the cellulose suspension in aqueous N-methylmorpholine-N-oxide produced is converted into the extrusion solution by water evaporation with shearing.

In contrast to known methods, in which the pulp is mixed directly with N-methylmorpholine-N-oxide (NMMO), in the method according to the present invention, a stronger activation and an increase in solubility of the cellulose is achieved by the aminoxide-free steps (a) to (c), so that the formation of solution in step (e) is accelerated and eased. Soluble attendant materials of the pulp may be washed out and partially removed from the method by the aminoxide-free steps (a) and (b) if only a part of the aqueous phase arising in step (b) is recycled in step (a)

and otherwise fresh water is used. Since, according to the present invention, thermal concentration of the cellulose suspension in aqueous NMMO before the step of solution formation is dispensed with, reduced thermal stress of the solution components results. The preferred mass ratio of cellulose/aqueous phase in step (a) is in the range from 1:10 to 1:30. The preferred duration of the shearing treatment of the suspension in step (a) is in the range from 10 to 120 minutes. The preferred cellulose content of the dewatered damp cellulose material in step (b) is in the range from 40 to 60 mass-percent.

According to the preferred embodiment, partly the aqueous phase from step (b) and partly fresh water is used for the production of the cellulose suspension in step (a). The remaining portion of the aqueous phase from step (b) is discarded. Soluble components carried along with the pulp are thus prevented from reaching the spinning solution in too high a proportion. On the other hand, the fine fiber component contained in the aqueous phase from step (b) is at least partially recycled and the loss of cellulose is thus minimized.

An aqueous phase is preferably used in step (a) which may contain soluble components, preferably up to 1 mass-percent.

In the preferred embodiment of the method according to the present invention, the cellulose suspension is dewatered with the aid of vacuum and/or pressure into a fleece and the water content of the fleece is determined with the aid of an infrared moisture measurement and the measured dimension is used to regulate the predetermined pressure parameter and/or the addition of the aqueous NMMO in step (d). Through this regulation, it is possible to continuously maintain the desired composition of the

extrusion solution, so that optimum properties of the extruded cellulosic molded bodies may be obtained.

The production of the cellulose solution is preferably performed in step (e) in a strong shear field with small heat exchange areas up to a NMMO/H_2O mol ratio in the range from 1:0.8 to 1:1.2. The energy necessary for water evaporation in this step is predominantly introduced into the viscous solution phase by shearing. In this way, and through the low addition of energy, localized overheating, and thus damage to the components of the extrusion solution, is avoided and the risk of the occurrence of exothermic reaction cycles is avoided. The water evaporation can also be regulated with less danger (runaway reaction) via the shear energy introduced than solely through the supply of heat via exchange surfaces.

In a separate embodiment of the method, the cellulose is enzymatically activated in step (a) or between steps (a) and (b) by treating the cellulose suspension with 0.01 to 10 mass-percent enzyme, in relation to the cellulose, at a temperature in the range between 20 and 70 °C, at a pH value in the range from 3 to 10, and for a duration in the range from 0.1 to 10 hours. The cellulose is more strongly activated for the dissolving procedure by this enzymatic treatment than by the shear treatment in step (a) alone. The dissolving of the cellulose in step (e) is thus accelerated; the cellulose solution formed is less viscous or is more concentrated at the same viscosity. Suitable enzymes (cellulases) are known in the related art, such as Rucolase from the firm Rudolph Chemie or Roglyr 1538 from the firm Rotta GmbH. The enzymatic treatment is particularly performed at 30 to 60 °C and at a pH value of 4.5 to 8 with 0.1 to 3.0 mass-percent cellulase during a period of 0.5 to 2 hours.

In a further embodiment of the method according to the present invention, at least steps (a) and (b) are performed following the pulp production in the pulp factory. These treatment steps may additionally include the enzymatic treatment. The performance of these steps during the pulp production has the advantage that the activation is possible with lower outlay than at the fiber or film producer, because apparatuses and experience in pulp treatment with aqueous media are available in the pulp factory. The fiber or film producer obtains a pulp activated according to his specifications, with which he can immediately begin step (c) of the method according to the present invention.

According to the present invention, the device for performing the method comprises a mixing tank with suspending elements, supply connecting pieces for pulp and aqueous suspension agent and drain connecting pieces for the suspension produced, a separating apparatus connected to the drain connecting pieces for partial separation of the suspension agent from the pulp, a return line for separated suspension agent, which leads from the separating apparatus to the supply connecting pieces for suspension agent on the mixing tank, a shearing apparatus, including a homogenization zone and an adjoining suspending zone, having a first feed connecting piece for pulp from the separation apparatus to the beginning of the homogenization zone, a second feed connecting piece for aqueous solvent at the beginning of the suspending zone, and a drain connecting piece for suspension at the end of the suspending zone, and a concentration and dissolving apparatus, having a feed connecting piece connected to the drain connecting pieces of the shear apparatus described, a solvent outlet connecting piece at the other end, and at least one vapor outlet connecting piece. This facility allows the continuous performance of the method. The individual apparatuses may be differently implemented in

this case. A suitable mixing tank for the suspending in aqueous suspension agent may be, for example, a pulper known in the related art. A suitable separating apparatus is preferably a vacuum screen belt press.

The present invention will now be described with reference to the drawing and the examples.

The figure schematically shows a facility for performing the method according to the present invention. A mixing tank 1 is fed pulp via connecting pieces 3. Suspension agent, comprising the aqueous recycled material supplied via a return line 4 and fresh water introduced via line 5, is supplied via connecting pieces 2. The aqueous pulp suspension formed in container 1 reaches a screen belt press 8 via line 7 from connecting pieces 6, on which it is dewatered down to a liquid content of 50 %. The suspension agent separated in this way, which carries along components dissolved from the pulp and fine fiber material, is recycled through return line 4 into mixing tank 1. A part of the recycled material may be discarded through line 9.

The dewatered pulp obtained on screen belt press 8 is supplied in fleece form via funnel 12 to a double shaft apparatus 11. In apparatus 11, multiple shafts having shear and conveyor elements are arranged, of which two shafts 13, 14 are illustrated in the drawing. In a first shear zone, which reaches over approximately the first third of the overall length of both zones, the shafts for the shearing of the aqueous cellulose introduced are set up. After approximately one-third of the apparatus length, there is a feed opening 15 for the solvent (aqueous NMMO) in the cylinder housing. Air and some water vapor is drawn off through lines 16. The apparatus is connected to the downstream end to a multishaft slusher 18, which is used as a dissolving station, by a tube 17. The suspension is transported by the superstructures of the shafts, with

shearing and dissolving of the cellulose, to output connecting pieces 19. Slusher 18 is kept under a partial vacuum by connecting pieces 20, which causes water to evaporate from the suspension and be drawn off from slusher 18. Both apparatuses 11 and 18 are provided with a heating mantle (not shown), so that the desired mixing and/or dissolving temperature may be maintained.

Example 1

70 kg pulp of the type MoDo with 6 % moisture was suspended in a pulper with 1300 l of completely desalinated water for approximately 10 minutes. A pulp suspension having 5 mass-percent pulp was produced. The suspension was fed at 50 °C and at a speed of 700 kg/h to a vacuum dewatering press, on which the pulp was dewatered to a moisture content of 50 %. 70 kg/h of the damp pulp was fed to the first shear zone of a shear apparatus schematically indicated in the figure. At the beginning of the second shear zone, 236 kg/h of aqueous N-methylmorpholine-N-oxide was introduced. The mixture was conveyed through the second shear zone, with the available apparatus cross-section being filled up essentially completely by the media conveyed. The slurry obtained had an NMMO content of 76.3 %. 306 kg/h of slurry was then dewatered further in an evaporator/slusher with a strong shearing field and small heat exchange surfaces until a homogeneous solution with an NMMO/H_2O mol ratio of 1:1 was formed. 270 kg/hour of spinning solution with a cellulose content of 12.3 % was obtained, which was drawn off from the aggregate at the temperature of 94.5 °C. The spinning solution could be assessed as good with reference to the refraction index, the particle content, the particle distribution in the solution, and its zero shear viscosity.

Example 2

70 kg of a pulp, which resulted in very poor spinning solution qualities under the method conditions of example 1, was whipped in a turbo slusher in water at 45 °C and a pH value of 7 in a bath ratio of 1:10, and treated with 1.5 mass-percent enzyme, in relation to cellulose, for 1.5 hours. The suspension formed was fed to a vacuum dewatering press at 50 °C and a speed of 700 kg/h. In this way, the pulp was dewatered to a moisture content of 50 %. The further processing of the fleece obtained was the same as an example 1. A spinning solution with good quality features was also obtained.

Example 3

34 kg/h of a pulp enzymatically pretreated in the pulp factory (0.5 % cellulase of the type Roglyr 1538 from the firm Rotta GmbH, in relation to cellulose, moisture content 6 %) was metered with the aid of a shredder having a discharge unit into a crusher via a belt weigher. The 34 kg/h was supplied with homogenization to a first shearing zone and, after the addition of 272 kg/h of 76 % NMMO, conveyed through a second shear zone. The resulting slurry had an NMMO content of 76.3 %. The slurry was processed further in the same way as in example 1. The spinning solution had the same good properties as in example 1.

CLAIMS

1. A method for the continuous production of an extrusion solution for the formation of cellulosic molded bodies, such as fibers and films, according to the lyocell method, wherein

   (a) a cellulose suspension is formed from pulp and an aqueous phase in a mass ratio in the range from 1:3 to 1:40 and maintained for a period of time in the range from 5 to 200 minutes with shearing,

   (b) the cellulose suspension is dewatered to form a material with a cellulose content in the range from 20 to 80 mass-percent and the aqueous phase resulting in this case is at least partially recycled in step (a),

   (c) the damp cellulose material is conveyed, with homogenization, through a first shear zone in the absence of N-methylmorpholine-N-oxide,

   (d) the homogenized cellulose material is conveyed through a second shear zone after the addition of sufficient aqueous N-methylmorpholine-N-oxide that after the mixing a suspension with a content of N-methylmorpholine-N-oxide in the liquid phase in a range from 70 to 80 mass-percent results, with the cellulose material filling up the available conveyor cross-section in the shear zones essentially completely, and

   (e) the cellulose suspension in aqueous N-methylmorpholine-N-oxide formed is converted into the extrusion solution by water evaporation with shearing.

2. The method according to Claim 1,

characterized in that

partly the aqueous phase from step (b) and partly fresh water is used for formation of the cellulose suspension in step (a).

3. The method according to Claim 1 or 2,

characterized in that

in step (a) an aqueous phase is used which may contain dissolved components.

4. The method according to one of Claims 1 to 3,

characterized in that

the cellulose suspension is dewatered in step (b) with the aid of vacuum and/or pressure into a fleece and the water content of the fleece is determined with the aid of an infrared moisture measurer and is used to regulate the predetermined pressure parameter and/or the addition of the aqueous N-methylmorpholine-N-oxide in step (d).

5. The method according to one of Claims 1 to 4,

characterized in that

in step (e) the formation of the cellulose solution is performed in a strong shear field with small heat exchange surfaces up to an NMMO/H_2O mol ratio in the range from 1:0.8 to 1:1.2.

6. The method according to one of Claims 1 to 5,

characterized in that

the cellulose is enzymatically activated in step (a) or between steps (a) and (b), by treating the cellulose suspension with 0.01 to 10 mass-percent enzyme, in relation to cellulose, at a temperature in the range between 20 and 70 °C and a pH value in the range from 3 to 10 for a duration in the range from 0.1 to 10 hours.

7. The method according to Claim 6,

characterized in that

the enzymatic treatment is performed with 0.1 to 3.0 mass-percent enzyme at 30 to 60 °C and a pH value of 4.5 to 8 for a duration of 0.5 to 2 hours.

8. The method according to one of Claims 1 to 7,

characterized in that

steps (a) and (b) are performed in the pulp factory.

9. A device for performing the method according to one of Claims 1 to 8, having

a mixing tank (1) with suspending organs, supply connecting pieces (3, 2) for pulp and aqueous suspension agent, and a drain connecting piece (6) for suspension,

a separating apparatus (8) connected with the drain connecting piece (6) for partial separation of the suspension agent from the cellulose,

a return line (4) for separated suspension agent from the separating apparatus (8) to a supply connecting piece (2) of the mixing tank (1) with a discard line (9) for the possible partial suspension agent discard,

a shearing apparatus (11), including a homogenization zone and an adjoining suspending zone, having a first feed connecting piece (12) for pulp from the separating apparatus (8) at the beginning of the homogenization zone, a second feed connecting piece (15) at the beginning of the suspending zone for solvent, and a drain connecting piece (17) for suspension at the end of the suspending zone, and

a concentration and dissolving apparatus (18) having a feed connecting piece connected to the drain connecting piece (17) of the shearing apparatus (11) at one end, a solution outlet connecting piece (19) at the other end, and at least one vapor outlet connecting piece (20).

10. The device according to Claim 9,

characterized in that

the separating apparatus (8) is a vacuum screen belt press.

11. The device according to Claim 9,

characterized in that

the separating apparatus (8) is a vacuum screen drum filter.

ABSTRACT OF THE DISCLOSURE

A method for the continuous production of an extrusion solution for the formation of cellulosic molded bodies, such as fibers and films, according to the lyocell method, in which (a) a cellulose suspension is formed from pulp and an aqueous phase in a mass ratio in the range from 1:3 to 1:40 and maintained for a period of time in the range from 5 to 200 minutes with shearing, (b) the cellulose suspension is dewatered to form a material with a cellulose content in the range from 20 to 80 mass-percent and the aqueous phase resulting in this case is at least partially recycled in step (a), (c) the damp cellulose material is conveyed, with homogenization, through a first shear zone in the absence of N-methylmorpholine-N-oxide, (d) the homogenized cellulose material is conveyed through a second shear zone after the addition of enough aqueous N-methylmorpholine-N-oxide that after the mixing a suspension with a content of N-methylmorpholine-N-oxide in the liquid phase in a range from 70 to 80 mass-percent results, with the cellulose material filling up the available conveyor cross-section in the shear zones essentially completely, and (e) the cellulose suspension in aqueous N-methylmorpholine-N-oxide formed is converted into the extrusion solution by water evaporation with shearing.

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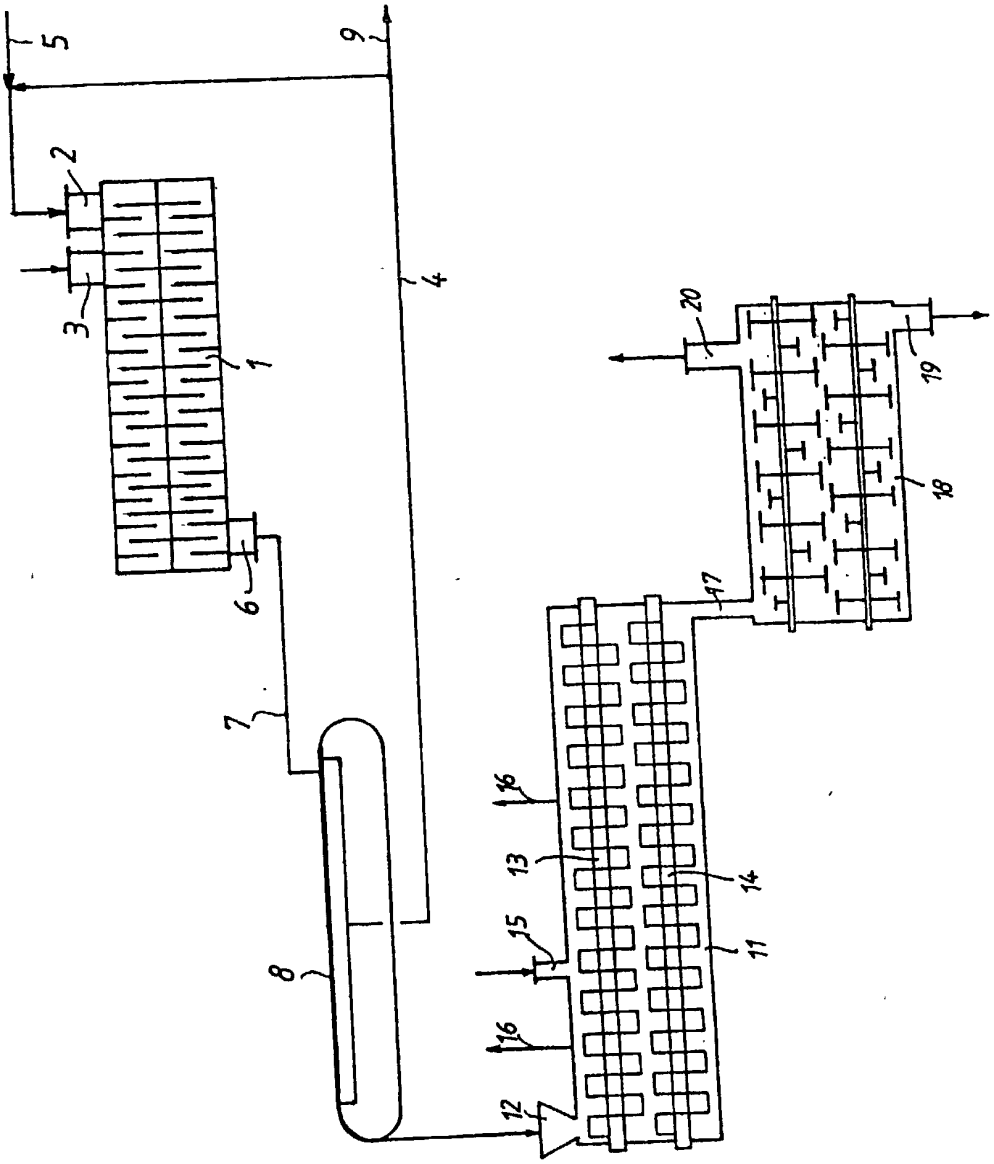

ALC-745

PATENT APPLICATION

DECLARATION AND POWER OF ATTORNEY FOR PATENT APPLICATION	**ATTORNEY DOCKET NO.** 4197-113

As a below named inventor, I hereby declare that:
My residence/post office address and citizenship are as stated below next to my name;
I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:
METHOD AND DEVICE FOR CONTINUALLY PRODUCING AN EXTRUSION SOLUTION
the specification of which is attached hereto unless the following box is checked:
(X) was filed March 18, 2002 as US Application Serial No. 10/088,773 or PCT International Application Number _____ and was amended on _____ (if applicable).
I hereby state that I have reviewed and understood the contents of the above-identified specification, including the claims, as amended by any amendment(s) referred to above. I acknowledge the duty to disclose all information which is material to patentability as defined in 37 CFR 1.56.

Foreign Application(s) and/or Claim of Foreign Priority
I hereby claim foreign priority benefits under Title 35, United States Code Section 119(a-d) or 365(b) of any foreign application(s) for patent or inventor(s) certificate, or 365(a) of any PCT international application which designated at least one country other than the United States of America, listed below and have also identified below any foreign application for patent or inventor(s) certificate having a filing date before that of the application on which priority is claimed:

COUNTRY	APPLICATION NUMBER	DATE FILED	PRIORITY CLAIMED UNDER 35 U.S.C. 119
Germany	199 49 720.6	15 October 1999	YES: X NO:
PCT	PCT/DE00/03411	29 September 2000	YES: X NO:

U.S. Priority Claim
I hereby claim the benefit under Title 35, United States Code, Section 120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code Section 112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, Section 1.56(a) which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

APPLICATION SERIAL NUMBER	FILING DATE	STATUS(patented/pending/abandoned)

POWER OF ATTORNEY:
As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) listed below to prosecute this application and transact all business in the Patent and Trademark Office connected therewith.

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

1-00

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Inventor's Signature **Date**

ACC-7/US

DECLARATION AND POWER OF ATTORNEY FOR PATENT APPLICATION (continued)

ATTORNEY DOCKET NO. 4197-113

2-00

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